

U.S. Patent Application Serial No. **10/508,954**  
Amendment filed February 15, 2006  
Reply to OA dated September 28, 2005

**REMARKS**

Claims 1 and 3-7 are pending. Claim 2 has been canceled.

Claim 1 has been amended with the contents of claim 2. The applicants respectfully submit that no new matter has been added. It is believed that this Amendment is fully responsive to the Office Action dated **September 28, 2005**.

**Claims 1-7 are rejected under 35 USC 102(b) as being anticipated by US 6,310,112, (Vo) see col.1-6 and examples. (Office Action p.2)**

US 6310112 B discloses a foam produced from a composition which comprises a hydrogenated vinyl aromatic polymer and may additionally comprise an additional polymer such as a polymer that contains lactic acid residuals (polylactic acid) and a polyester (see claims 1 and 11). The composition of US 6310112 B is a molten mixture. *The molten mixture is clearly different from a block copolymer produced by reacting a polylactic acid and a polyester in accordance with the claimed invention.* Therefore, the composition disclosed in US 6310112 B is outside of the technical scope specified in Claim 1 of the present application, in which the block copolymer is an essential component.

In addition, the molten mixture disclosed in US 6310112 B may not have a microphase separated structure stipulated in Claim 1 of the present application, in the same manner as demonstrated in Comparative Example 3 (molten mixture) of the specification of the present application as originally filed.

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Consequently, the invention in accordance with the amended Claim 1 of the present application is not described in or even suggested by US 6310112B. Since US 6310112B nowhere discloses *reacting a polylactic acid and a polyester*, it cannot legally anticipate the invention as now claimed. Additionally, the invention in accordance with the original Claims 3 to 7 of the present application is also not anticipated by US 6310112B.

It is respectfully requested that the rejection be favorably reconsidered and withdrawn.

**Claims 1-7 are rejected under 35 USC 102(e) as being anticipated by US 6,756,331, see abstract, col.1-4, table 3, claims 2 and 4. (Office Action p.5)**

US 6756331 B discloses a lactic acid-based resin composition containing a mixture of:

(A) a mixture of:

(a1) a polylactic acid and

(a2) an aliphatic polyester, and

(B) an aliphatic block co-polyester having a polylactic acid segment and an aliphatic polyester segment.

*The mixture (A) recited in claims 2 and 4 of US 6756331 B is a simple mixture which is clearly different from a block copolymer in accordance with the claimed invention.* Therefore, the mixture (A) disclosed in US 6756331 B may not have a microphase separated structure stipulated in Claim 1 of the present application, as demonstrated in Comparative Example 3 (molten mixture) of the specification of the present application as originally filed.

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In addition, the aliphatic block co-polyester (B) is produced by, for example, reacting polybutylene succinate and polylactic acid in an organic solvent such as diphenyl ether (see Production Example 59 in Column 12 of US 6756331 B). *When a polyester and a polylactic acid are reacted in an organic solvent to obtain a molding resin, the obtained molding resin fails to maintain the desired storage elastic modulus with respect to strain, as specified in the amended Claim 1* of the present application. Therefore, the molding resin formed in accordance with US 6756331 B is chemically different from the molding resin stipulated in the amended Claim 1 of the present application. This is demonstrated empirically in the attached Inventor's Declaration.

In addition, US 6756331 B fails to describe or suggest that in the case of reacting a polyester and a polylactic acid *without an organic solvent*, a molding resin maintaining the desired storage elastic modulus with respect to strain can be obtained.

Consequently, the invention in accordance with the amended Claim 1 of the present application is not described or suggested in US 6756331 B. Because of the stated chemical difference over the reference, it cannot possibly anticipate claim 1. Additionally, the invention in accordance with the original Claims 3 to 7 of the present application is not described in or suggested by US 6756331 B.

On this basis, it is respectfully requested that the rejection be favorably reconsidered.

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**Claims 1-3 are rejected under 35 USC 102(e) as being anticipated by EP 712 880B, see abstract and pages 2-4. (Office Action p.7)**

EP 712880 B discloses that a polyester prepared from a diol and a dicarboxylic acid is reacted with a polylactic acid in the presence of a catalyst in an organic solvent (see [0048], [0057], and the like of EP 712880 B).

As described above, in the case of preparing a molding resin by reacting a polyester and a polylactic acid *in an organic solvent*, i.e., by subjecting a polyester and a polylactic acid to an azeotropic dehydration polycondensation reaction in an organic solvent, *the obtained molding resin fails to maintain the desired storage elastic modulus with respect to strain, as specified in the amended Claim 1 of the present application*. Therefore, the molding resin formed in accordance with EP 712880 B is chemically different from the molding resin stipulated in the amended Claim 1 of the present application. This is demonstrated empirically in the attached Inventor's Declaration.

In addition, EP 712880 B fails to describe or suggest that in the case of reacting a polyester and a polylactic acid *without an organic solvent*, a molding resin maintaining the desired storage elastic modulus with respect to strain can be obtained.

Consequently, the invention in accordance with the amended Claim 1 of the present application is not described in or suggested by EP 712880 B. Because of the stated chemical difference over the reference, it cannot possibly anticipate claim 1. Additionally, the invention in accordance with the original Claims 3 to 7 of the present application is not described or suggested in EP 712880 B.

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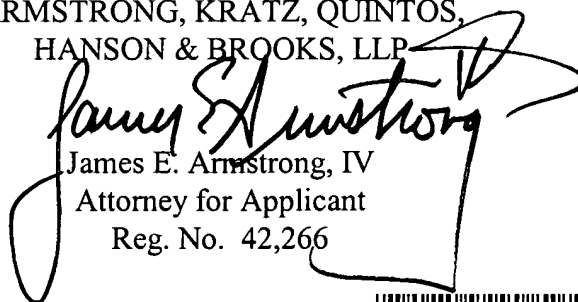
In view of the aforementioned amendments and accompanying remarks, the claims, as amended, are in condition for allowance, which action, at an early date, is requested.

If, for any reason, it is felt that this application is not now in condition for allowance, the Examiner is requested to contact the applicants undersigned attorney at the telephone number indicated below to arrange for an interview to expedite the disposition of this case.

In the event that this paper is not timely filed, the applicants respectfully petition for an appropriate extension of time. Please charge any fees for such an extension of time and any other fees which may be due with respect to this paper, to Deposit Account No. 01-2340.

Respectfully submitted,

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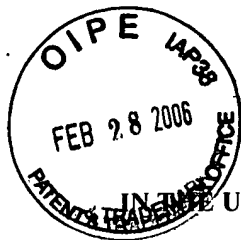
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PATENT TRADEMARK OFFICE

Enclosures: Declaration  
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UNITED STATES PATENT AND TRADEMARK OFFICE

In re the Application of: **MIHARA, Takashi, et al.**

Group Art Unit: 1711

Serial No.: 10/508,954

Examiner: **Terressa M. BOYKIN**

Filed: **October 4, 2004**

**P.T.O. Confirmation No.: 7737**

For. **RESIN FOR MOLDING AND PRODUCTION PROCESS THEREFOR**

**DECLARATION UNDER 37 CFR 1.132**

Commissioner for Patents

P.O. Box 1450

Alexandria, VA 22313-1450 February 27, 2006

Sir:

I, Takashi MIHARA, a citizen of Japan, hereby declare and state as follows:

**I. Background**

1. I have a master's degree in Industrial Chemistry which was conferred upon me by the Graduate School of Science and Technology at the Nihon University in Japan, in March 1998.

2. I have been employed by DAINIPPON INK AND CHEMICALS, INC., since April 1998, and have been engaged in research in specialty chemical areas.

3. I am a member of the Plasticizer & Modifier Group of Additives & Chemicals in DAINIPPON INK AND CHEMICALS, INC.

4. My publications include the following works in this field:

*Masao Kamikura, Shouji Imamura, Akio Toyoda, and Takashi Mihara, DIC Technical Review, No. 10 (2004).*

I carried out the following Experiments described below.

## II. Experiments

### Reference Example 1: Synthetic method for an aliphatic polyester

In a reaction vessel, were placed 527 g of propylene glycol (manufactured by Asahi Glass Co., Ltd.) (hereafter abbreviated as PG) and 1,000 g of sebacic acid (manufactured by Kokura Synthetic Industries, Ltd.) (hereafter abbreviated as SeA), and with a stream of nitrogen passing through the vessel, the temperature was increased, with stirring, from 150°C to 230°C at a rate of 10°C per hour, while the generated water was removed, thus effecting an esterification reaction. After the reaction was continued for one hour at 230°C, titanium tetrabutoxide (manufactured by Kishida Chemical Co., Ltd., the first grade reagent) was added as a polymerization catalyst in a quantity equivalent to 70 ppm relative to the weight of the raw materials, and the pressure was then reduced to 200 Pa to react with heating and stirring. Eight hours after starting the reduction of the pressure, a GPC measurement was carried out. As a result, an aliphatic polyester with a number average molecular weight (hereafter abbreviated as Mn) of 32,000 and a weight average molecular weight (hereafter abbreviated as Mw) of 58,000 was obtained. In addition, 2-ethylhexanoic acid phosphate was added as a deactivation agent for the polymerization catalyst, in a quantity equivalent to 80 ppm relative to the aliphatic polyester, and deactivation of the polymerization catalyst was carried out for one hour. The molecular weight of the obtained aliphatic polyester (B) of PG-SeA was measured by means of GPC. As a result, Mn was 33,000, and Mw was 58,000.

**Experiment [1]. Production of a block copolymer in an organic solvent (corresponding to the cases of Production Example 59 of US 6756331 B and [0048] of EP 712880 B with the proviso that the block copolymer is not a polybutylene succinate-based copolymer)**

In a reaction vessel, were placed 25 g of the aforementioned aliphatic polyester (B), 25 g of polylactic acid (manufactured by Mitsui Chemicals Inc., LACEA H400) (hereafter abbreviated as PLA (A)), tin powders as a catalyst in a quantity equivalent to 4,000 ppm relative to the weight of the PLA (A) and the aliphatic polyester (B) in total, and 450 mL of diphenyl ether. The mixture was reacted for 40 hours at 160°C/10 mmHg, with heating and stirring, in order to avoid contamination of water in the reaction system by fitting a tube which is charged with 20 g of molecular sieves 3A (manufactured by Wako Pure Chemical Industries, Ltd.) to the reaction vessel so that the solvent distilled due to refluxing returned into the reaction system through the molecular sieves. The obtained molding resin was soaked in methanol, and subsequently soaked in hexane. The soaking

operation was repeated 5 times in total. Thereby, diphenyl ether included in the molding resin was removed. The molding resin in which diphenyl ether was removed was dried under reduced pressure for 5 hours at 90°C. The molecular weight of the dried molding resin (P-1) was measured by means of GPC. As a result, Mn was 49,000, and Mw was 95,000. In addition, thermal properties of the obtained molding resin were measured by means of a differential scanning calorimeter (DSC) (Q 100 model, manufactured by TA Instruments Inc.). As a result, the glass transition temperatures (Tg) were observed at -52°C and 60°C, and the melting points (Tm) were observed at -26°C and 165°C.

In order to confirm the microphase separated structure of the obtained molding resin, the morphology of the aforementioned molding resin was observed by means of a transmission electron microscope (TEM). As a result, it could be seen that domains which the structural units of PLA (A) formed were formed in a matrix which the structural units of aliphatic polyester (B) formed. In addition, the average domain size of the aforementioned domains ranged from about 0.2 to 3 µm.

#### **Experiment [2]. Production of a molding resin in accordance with the present invention**

In a reaction vessel filled with a nitrogen gas, was placed 25 g of the aforementioned aliphatic polyester (B), and was subjected to heat-melting at 210°C. Subsequently, 25 g of PLA (A) was added thereto, and the mixture was subjected to molten mixing at 210°C. After the aforementioned aliphatic polyester (B) and the PLA (A) had been visually confirmed as having formed a uniform molten mixture, titanium tetrabutoxide was added as an esterification catalyst, in a quantity equivalent to 200 ppm relative to the total of the aforementioned molten mixture. The reaction was allowed to proceed for 5 hours under reduced pressure of 80 Pa. After completion of the reaction, 2-ethylhexanoic acid phosphate was added as a deactivation agent for the esterification catalyst, in a quantity equivalent to 400 ppm relative to the total amount of the obtained molding resin. The mixture was reacted for one hour under reduced pressure, and thereby the esterification catalyst was deactivated. The molecular weight of the obtained molding resin (P-2) was measured by means of GPC. As a result, Mn was 65,000 and Mw was 123,000. In addition, the thermal properties of the obtained molding resin were measured by means of a differential scanning calorimeter (DSC) (Q 100 model, manufactured by TA Instruments Inc.). As a result, the glass transition temperatures (Tg) were observed at -60°C and 55°C, and the melting points (Tm) were observed at -27°C and 165°C.

In order to confirm the microphase separated structure of the obtained molding resin, the morphology of the aforementioned molding resin was observed by means of a TEM. As a result, it



could be seen that domains which the structural units of aliphatic polyester (B) formed were formed in a matrix which the structural units of PLA (A) formed. In addition, the average domain size of the aforementioned domains ranged from about 0.2 to 3  $\mu\text{m}$ .

### III. Results

#### A. Measurement of storage elastic modulus

With respect to the aforementioned molding resin (P-1) obtained in Experiment [1] and the aforementioned molding resin (P-2) obtained in Experiment [2], the storage elastic modulus  $G'$  was measured by means of an ARES viscoelasticity measurement device (rotational rheometer) manufactured by TA Instruments Inc., when the strain was changed from 1% to 60% (see Fig. 1). The measurement conditions were as follows. Frequency: 1 Hz, measurement temperature: 180°C, jig: parallel plates (diameter 25 mm), and gap (the distance between the parallel plates): 1 mm.

With respect to the aforementioned molding resin (P-1) obtained in Experiment [1] and the aforementioned molding resin (P-2) obtained in Experiment [2], the values for the storage elastic modulus  $G'$  (1%) when the strain was 1% and the storage elastic modulus  $G'$  (60%) when the strain was 60% were read based on Fig. 1, and the ratio  $(100 \times G' (60\%) / G' (1\%))$  between the values  $G'$  (60%) and  $G'$  (1%) was calculated (see Table I).

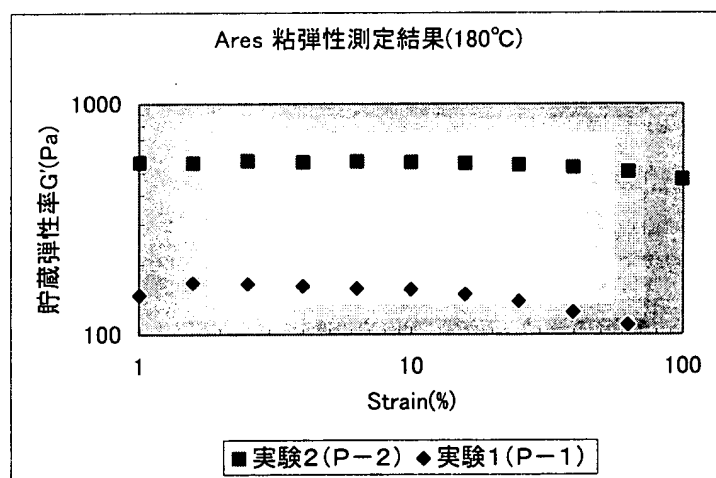


Fig. 1 Relationship between strain and storage elastic modulus of each of molding resin (P-1) and molding resin (P-2)

In Fig. 1,

“Ares 粘弾性測定結果 (180°)” =

ARES viscoelasticity measurement results (180°C),

“貯蔵弾性率  $G'$ (Pa)” = Storage elastic modulus  $G'$  (Pa),

“実験 2 (P-2) ” = Experiment [2] (P-2)

“実験 1 (P-1) ” = Experiment [1] (P-1)

Table I (G' (60%) /G' (1%)) X 100

	Experiment [1]	Experiment [2]
Molding resin	(P-1)	(P-2)
(G' (60%) /G' (1%)) X 100	75%	92%

As described above, molding resin (P-1) (a comparative molding resin obtained by a reaction between a polyester and a polylactic acid in an organic solvent in accordance with Production Example 59 of US 6756331 B and [0048] of EP 712880 B) in which (G' (60%) /G' (1%)) X 100 is 75%, is clearly different from molding resin (P-2) in accordance with the present invention, in which (G' (60%) /G' (1%)) X 100 is 92% and is within a range of 90 to 100%, stipulated in the amended Claim 1. Therefore, it can be seen that in a molding resin obtained by a reaction between a polyester and a polylactic acid in an organic solvent, a storage elastic modulus G' (M%) at a strain of M% ( $1 < M \leq 60$ ) cannot be maintained within a specified range of from 90 to 100% of a value of a storage elastic modulus G' (1%) at the strain of 1%. In contrast, in a molding resin obtained by an esterification reaction between a polyester and a polylactic acid without organic solvents, a storage elastic modulus G' (M%) at the strain of M% ( $1 < M \leq 60$ ) can be maintained within a specified range of from 90 to 100% of a value of a storage elastic modulus G' (1%) at the strain of 1%, as stipulated in the amended Claim 1 of the present application.

Consequently, the invention stipulated in each of Claims 1 and 3 to 5 is not described in or suggested by US 6756331 B and EP 712880 B.

## **B. Evaluation of physical properties**

### **1. Preparation methods of polyester composition and film thereof**

PLA (A) and the molding resin (P-1) were dried under reduced pressure for 3 hours at 70°C. Subsequently, using the mixing ratios indicated in Table II, the materials were subjected to molten mixing for 10 minutes at 190°C using a laboplast mill mixer manufactured by Toyo Seiki Kogyo Co., Ltd., thus forming polyester compositions (Q-1) and (Q-2). In addition, in the same manner as described above, PLA (A) and the molding resin (P-2) were subjected to molten mixing, thus forming polyester compositions (Q-3) and (Q-4).

Each of the obtained polyester compositions was dried under reduced pressure for 3 hours at 70°C. Subsequently, a thermal press device at a temperature of 195°C was used to prepare films having a thickness of 200  $\mu\text{m}$ .

## 2. Method of measuring transparency

The transparency of each of the films obtained by molding in accordance with the aforementioned method was evaluated by carrying out haze measurements using a turbidity meter (ND-1001DP, manufactured by Nippon Denshoku Industries Co., Ltd.). The lower the haze value is, the better transparency is.

## 3. Method of testing Dupont impact strength

Using a Dupont impact strength measurement apparatus, the 50% rupture energy of each of the aforementioned films was obtained in accordance with JIS K 5400.

Table II: Evaluation results of physical properties of polyester compositions

		Comparative Experiment Example 1	Comparative Experiment Example 2	Experiment Example 1	Experiment Example 2
Polyester composition		Q-1	Q-2	Q-3	Q-4
PLA (A)	parts by weight	90	70	90	70
Molding resin (P-1)		10	30	-	-
Molding resin (P-2)		-	-	10	30
Haze value (%)		11	46	9	33
Dupont impact strength (mJ)		110	380	350	800

## IV. Conclusion

As is clear from the results of Table II, the physical properties of the polyester compositions of Experiment Examples 1 and 2 in accordance with the present invention as claimed in the amended Claim 1 of the present application exhibited better overall evaluation, compared to the physical

properties of the polyester compositions of Comparative Experiment Examples 1 and 2, respectively.

This empirical data shows that the invention according to each of Claims 6 and 7 of the present application is not, in fact, described in or suggested by US 6756331 B and EP 712880 B.

The undersigned declares that all statements made herein of his/her own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under ' 1001 of Title 18 of the United States Code and that willful false statements may jeopardize the validity of the application or any patent issued thereon.

Signed this 15<sup>th</sup> day of February, 2006

Takashi, MIHARA

*Takashi Mihara*

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